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NEW USSR DATA ON THE MECHANISM OF CHEMICAL REACTIONS
IN THE SOLID PHASE AND THEIR SIGNIFICANCE
FOR THE PREPARATION OF CATALYSTS

S. Z. Roginskiy

It is customary in chemical kinetics to regard the majority of reactions occurring in the solid phase as instances of a phenomenon which may be described as chemical crystallization. The character and rate of this crystallization depend on relationships between the rate of nuclei formation of crystals of the new phase, the rate of growth of the newly formed nuclei, and the spatial distribution of these processes. However, many experimental facts and recently established relationships are incompatible with the concept of crystallization.

An investigation of the mechanism and spatial distribution of reactions of this type was carried out by a method combining structural studies with crystallographic and adsorption measurements. This investigation comprised observations on the dehydration of crystal hydrates, the elimination of ammonia from ammoniates, the decomposition of heavy metal carbonates, the decomposition of solid permanganates, and the photochemical decomposition of silver bromide.

The investigation in question demonstrated that formation of crystals of the new phase is not a primary process occurring as a result of the reaction; this process is preceded by several important stages which hitherto have escaped the attention of investigators. The first stage consists in the removal of atoms and molecules from the lattice of the solid body, so that molecular porosity results. At the same time, pseudomorphism with the crystals of the starting material is retained. In some cases, initial retention of the original structure on formation of porous products can be demonstrated by X-ray investigation. Subsequent X-ray and electron-microscopic examinations disclose complete transition into the amorphous state. The next stage is enlargement of the pores and appearance of a skeleton structure which can be observed under an electron microscope. This process is succeeded by, and occasionally accompanied by, crystallization of the amorphous substance, this crystallization being produced by movement of atoms in the lattice through diffusion.

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Depending on the bond strength with which individual atoms or groups are held in the lattice and the dimensions of these atoms and groups, the processes described above may stop at any of the stages which have been mentioned. Thus, in a number of silicates and silicate-ammine systems, the process stops at the molecular skeleton stage. This is the zeolite type of process which leads to active adsorbents, catalyst carriers, and catalysts having an ultraporous surface. On the other hand, if the reaction products are metals, the process cannot be easily stopped in the initial stages, and skeleton structures with large pores which are visible under the electron microscope result immediately. Furthermore, the walls of these skeleton structures exhibit crystallinity under X-ray examination. In the case of oxidation-reduction reactions, all transformations indicated above are preceded by electron transitions in the solid phase.

The results obtained in the investigation outlined above facilitate application of topochemical reactions in connection with the production of active adsorbents having a certain set of desired properties. The intermediate formation of molecularly porous and colloiddally porous substances is important from the viewpoint of capture of various admixtures and additives by the substances in question. This is a phenomenon of essential significance in the production of catalysts for practical use. An understanding of the underlying relationships may also be of help in the preparation of luminophores.

The results of the investigation demonstrate a fundamental unity of the primary processes of decomposition and phase transformation of solid bodies, clarified in the course of the work in question, with the phenomena of exchange, combination, and recrystallization generally occurring in building materials, alloys, and ceramics. Processes of this type must also play an important role in the fields of geochemistry and mineralogy.

According to FDD Summary No 28, Vol I, S. Z. Roginskiy, Corresponding Member, Academy of Sciences USSR, is chief of the Nickel Catalysis Section, Laboratory of the Structure of Surface Films, Institute of Physical Chemistry (Moscow), Academy of Sciences USSR. He presented the paper reported on here at the regular meeting of the Division of Chemical Sciences of the Academy which took place on 28-29 March 1950.

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